METHOD OF SEPARATING A SOLID COMPOUND FROM AN ADDUCT

Related Applications

This patent application is a continuation-in-part application of U.S. Patent

5   Application Serial No. 10/099,227 (Attorney Docket no. A-71155/MSS) filed on March

13, 2002, the entire disclosure of which is incorporated herein by reference.

Field of the Invention

The present invention relates in general to a method of separating one or more

10   solid compound from an adduct. More specifically, the present invention provides a

method of separating one or more solid compounds from an adduct by crystallization

wherein certain phase equilibrium conditions are selectively controlled.

Background of the Invention

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Separation technologies are often used in the chemical industry to separate or

purify chemical compounds. One widely employed separation technology is

crystallization, wherein one or more solid compounds are formed from a solution.

Crystallization techniques, such as melt crystallization and solution crystallization, and

20   the associated unit operations, are well known in the industry.

Crystallization becomes more complex when a single solid component is not

easily formed from the solution, but instead an adduct is formed. An adduct is a

compound formed by direct combination of two or more different compounds or elements. The adducts are typically formed in simple molecular proportions, such as an equal molar combination. Formation of the adduct is undesirable, since it is one or more of the single compounds that is the product of interest.

5   When an adduct of two components is formed, wherein only one of the two components is the desired product, a variety of recovery processes are used in prior art techniques to obtain the desired product. Most techniques center around driving off the undesired component, such as by melting the adduct and then removing the undesired components by distillation, and other like means. The adduct is often first purified by

10   multiple steps of washing the adduct, dissolving the adduct, and re-crystallizing the adduct. All such processes are quite costly and energy intensive.

Accordingly, there is an important need in the industry to provide a method of separating a compound from an adduct which is free of such undesirable steps. Direct separation of a compound of interest would be a significant advance in the industry.

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Summary of the Invention

Accordingly, the present invention provides a method of separating one or more solid compounds from solution where the one or more solid compounds may form an

20   adduct. In particular, the present invention provides a method of separating one or more solid compounds or products from an adduct by crystallization wherein certain phase equilibrium conditions are selectively controlled to form either one or more of the desired

solid compounds or an adduct. By forming a desired solid compound directly, the costly and complex recovery steps of the prior art are avoided. By providing a method that allows one to specifically select the formation of one or more of the desired solid compounds, or the adduct, a powerful process development tool is created, whereby

5   certain operating conditions or variables of a process can be selectively adjusted to generate the desired phase equilibrium behavior.

In one embodiment, the present invention provides a method of separating one or more solid compounds from a solution including one or more components in a process operation, and where the one or more components may form at least one adduct,

10   characterized in that: at least two solvents are provided in the solution, at least a first solvent exhibiting a first phase behavior and at least a second solvent exhibiting a second phase behavior, and the concentration of the solvents are selectively controlled such that a solid compound is formed from the solution at selected process operating conditions.

In another embodiment of the present invention, a method of manipulating a

15   phase equilibrium behavior of a solution having two or more components and which may be represented by a multi-component phase diagram is provided, characterized in that:

the solution is comprised of multiple components, including at least two components, and a solvent component, and where the at least two components may form an adduct;

20   and where the multiple components exhibit a phase equilibrium behavior which can be represented by a multi-component phase diagram and where the components are

in phase equilibrium and exhibit phase behavior which establishes at least two regions in the phase diagram, an adduct region and at least one pure solid compound region; and

wherein the solvent component is comprised of two or more solvents, at least a first solvent exhibiting a first phase behavior and at least a second solvent exhibiting a

5   second phase behavior, and the concentration of the solvents in the solution are adjusted to selectively manipulate the area of one or more of the at least two regions.

In yet another aspect, the present invention provides for manipulating the relative size of the adduct and pure compound component regions by adjusting the concentration ratio of the two or more solvents; adjusting the composition of the solution prior to

10   crystallization to place the solution concentration at a location selectively within either the adduct or one of the pure compound component regions; and crystallizing the solution to form the desired compound from the solution.

<u>Brief Description of the Drawings</u>

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Other objects and advantages of the present invention become apparent upon reading of the detailed description of the invention provided below and upon reference to the drawings in which:

FIG. 1A is a solid-liquid phase equilibrium diagram for a system in which

20   components A and B react to form an adduct C.

FIG. 1B is a projection of the phase diagram of the binary system of FIG 1A along the temperature axis.

FIG. 2 is a projection of a poly-thermal phase equilibrium diagram for a ternary system comprising the components A, B and their adduct C, along with a solvent.

FIGs. 3 and 4 are projections of a poly-thermal phase equilibrium diagrams for the system of Figure 2 illustrating the case where the solvent compartment becomes very small when the melting temperature of the solvent is much lower than the melting temperature of both components A and B.

Fig. 5 is a projection of a poly-thermal phase equilibrium diagram for a multi-component system of components A, B, an adduct C formed from A and B, and Solvent I, showing the phase regions as a function of the solvent composition according to one embodiment of the present invention.

Fig. 6 is a projection of a poly-thermal phase equilibrium diagram for a multi-component system of components A, B, an adduct C formed from A and B, and solvent II showing the phase regions as a function of the solvent composition according to another embodiment of the present invention.

FIGs. 7 and 8 are projections of a poly-thermal phase equilibrium diagrams for a multi-component system of components A, B and an adduct C formed from A and B, illustrating an overlap region created by the selection of Solvent I and Solvent II (collectively referred to as "Solvent") according to another embodiment of the present invention.

FIG. 9 is a projection of a poly-thermal phase equilibrium diagram for a multi-component system of A, B, an adduct C formed from A and B, and solvent showing

maximum recovery of component A using two different solvents, at a specific feed composition to a crystallizer according to one embodiment of the present invention.

FIG. 10 is a schematic process block diagram of a separation system which may be used to carry out the method of one embodiment of the present invention.

FIG. 11 is a projection of a poly-thermal phase equilibrium diagram for a multi-component system corresponding to the separation system of FIG. 10.

FIG. 12 is a schematic process block diagram of a separation system which may be used to carry out the method of another embodiment of the present invention.

FIG. 13 is a projection of a poly-thermal phase equilibrium diagram for a multi-component system corresponding to the separation system of FIG. 12.

FIG. 14 is a projection of a polythermal phase equilibrium diagram for a multicomponent system of Example 1, showing the effect of solvent compositions in the crystallizers, on the overlap between the pure component phase regions.

FIGs. 15-17 are projections of a poly-thermal phase equilibrium diagram illustrating the stream compositions for three different cases for the separation flowsheet described in Example 2.

FIGs. 18-19 are projections of a poly-thermal phase equilibrium diagram illustrating the stream in the separation flowsheet described in Example 3 for two different solvent compositions.

FIG. 20 is a projection of a poly-thermal phase equilibrium diagram illustrating the stream compositions in the above mentioned separation flowsheet when utilizing the "swing" in the solvent composition from Solvent I to Solvent II.

Detailed Description of the Invention

The inventors have discovered a method of separating one or more solid

5   compounds from a solution an adduct by crystallization where the one or more solid

compounds may form at least one adduct. Certain phase equilibrium conditions are

selectively controlled to form either the one or more desired solid product(s) or an

adduct. By forming the desired solid compound(s) directly, the costly and complex

recovery steps of the prior art are avoided. By providing a method that allows one to

10   specifically select the formation of either the desired solid compound or an adduct, a

powerful process development tool is created, whereby certain operating conditions or

variables of a process can be selectively adjusted to generate desired phase equilibrium

behavior. The invention provides for manipulation and selective control of the phase

equilibrium of a multi-component liquid/solid phase equilibrium system. Typically the

15   multi-component phase equilibrium system will be directed to a ternary or quaternary

system; however the method of the present invention may be applied to a system with

any number of components.

In general, the present invention is directed to multi-component phase equilibrium

systems, where at least two of the components form an adduct. For example, the system

20   may be comprised of a solution of components A and B, and such components form an

adduct C. Either component A or B, or in some instances both A and B, may be the

compound or product which one wishes to separate from the solution. To illustrate the principles of the invention, exemplary phase diagrams are used as described below. Exemplary phase diagrams are shown which have certain characteristics solely in order to illustrate the invention, and the invention is not to be construed to be limited in any way

5   to the specific examples shown.

Referring to Fig. 1A, a phase diagram for a binary system in which component A and B form an adduct C is shown. The system is characterized as a solid and liquid equilibrium with the presence of a congruently melting solid compound. The maximum in the solid and liquid equilibrium results from the formation of a solid addition

10   compound C, which is equi-molar in A and B and with the molecular formula A-B, that melts at the temperature corresponding to the maximum, to form a liquid with the same composition. This type of behavior is well known in the art as congruent melting. On the phase diagram the x axis represents the mass concentration of component B and the y axis is the temperature.

15   In this example two eutectic points, E_1 and E_2 are present, and five separate phase regions are characterized that are important for recovery of solids according to the present invention: Region I where components A and B are in the liquid phase; Region II where solid A and a liquid mixture of A and B are in equilibrium. Region III and IV where solid C (adduct) and a liquid mixture of A and B are in equilibrium; and Region V

20   where solid B and a liquid mixture of A and B are in equilibrium. An additional region below the eutectic point E1 exists where solid A and solid C are present, and below

eutectic point E2 where a mixture of solid C and solid B are present; however these regions are not treated further here as recovery of only a pure solid is of interest.

Figure 1B shows the projection of this poly-thermal diagram along the temperature axis. For a binary system as the one under consideration, the projection

5   becomes a line, A–B, and the locations of E_1 and E_2 are placed along the line as shown in the figure. It should be noted that the location of the congruent melting point C falls between the two eutectic points E1 and E2.

The inventors have discovered that this projection of the binary poly-thermal diagram is useful for process synthesis because the recovery of each solid can be

10   visualized from this projection. Referring to Figure 1A, point F represents the feed composition of the solution at a specific temperature, which in this example places the location of point F in Region III. The recovery of solid adduct C, which can be obtained from this composition can be found from the polythermal projection shown in Figure 1B. Using the well known lever rule, the recovery of solid adduct C is the ratio of FM to CM.

15   The maximum recovery of pure solid C from this feed is the ratio of FE_1 to CE_1, because both solid C and solid A coexist when the temperature drops to eutectic point E_1.

Similarly, as known in the field, for a starting composition located in any of the other phase regions, the identity of the solid recovered, as well as its maximum recovery can be determined from the projection of the polythermal phase diagram. For point F

20   located in region I, the maximum recovery of A is given by the ratio of FE_1 to AE_1. For point F located in region IV, the maximum recovery of C is given by the ratio of FE_2 to

CE_2. For point F located in region V, the maximum recovery of B is given by the ratio of FE_2 to BE_2.

According to the method of the present invention, the poly-thermal diagram can be employed as a useful tool for process design and feasibility studies because it

5   identifies the region where the desired product can be recovered. The projection of the poly-thermal diagram becomes very useful when the complete phase diagram is 3-dimensional or 4-dimensional. Of particular advantage the process alternatives for separating the desired pure components can be visualized according to the present invention.

10   The present invention is now described in more detail. The inventors have discovered a method of separating one or more solid compounds from a solution including a solvent and multiple components and where at least two of the components form one or more adducts, by selectively controlling the phase equilibrium conditions through manipulation of the solvent composition and/or concentration in the solution.

15   This is a very powerful tool and allows one to generate a separation process with process operating conditions such as, but not limited to, type of solvent(s); concentration ratio of solvents when two or more solvent components are employed; or the mass concentration of any one of, or combination of, the components fed to a crystallizer.

In an exemplary embodiment, the system is comprised of three components: A

20   and B and a solvent S. Components A and B form an adduct C. Component A and/or B and/or the adduct C may be the compound of interest for separation from the solution. The isobaric phase diagram of the ternary system represents an image in 3-dimensional

space. The composition coordinates can be plotted in mole fractions on a triangular grid, and the temperature can be plotted on an additional axis. However, it is not convenient to work with multi-dimensional phase diagrams. Fortunately, for this three component system, much of the important information, particularly the crystallization boundaries,

5   can be represented on a 2-dimensional projection onto a triangular base. This is referred to as the projection of a poly-thermal phase equilibrium diagram. Please note that the ternary phase equilibrium diagrams are represented here as right-angled triangular diagrams for clarity as opposed to the alternative equilateral triangular diagram form also known in the art. While a three component system is described, it should be understood

10   by those of ordinary skill in the art that the system may include other components or solutes such as impurities, unreacted reactants, isomers and the like; however, for purposes of describing the present invention we are concerned only with the primary components in the solution.

To illustrate the system and method of the present invention, and in particular the

15   selective control of phase equilibrium conditions, first the phase diagrams are established and described. Referring to the Fig. 2, in the exemplary embodiment four phase regions or compartment are identified in the projection of the polythermal phase diagram for the ternary system of A, B and solvent. Components A and B may form an adduct C. When the feed to the crystallizer is located in compartment A, pure solid A component is

20   recovered. Compartment C is where a solid adduct of A and B, called adduct C is recovered. Compartment B is where pure solid B is recovered. A solvent compartment is present where solid solvent S may be recovered.

In this example four binary eutectics are present: E_1, E_2, E_{SA} and E_{SB}. E_1 is the eutectic point between component A and component C. E_2 is the eutectic point between component C and component B. E_{SA} is the eutectic between solvent S and component A, and E_{SB} is the eutectic point between solvent S and component B.

5   In this exemplary embodiment, two ternary eutectic are present, namely E_{SAC} and E_{SBC}. E_{SAC} represents a ternary eutetic of solvent, component A and component C. E_{SBC} represents the ternary eutetic of solvent, component B and component C. The trajectory of the eutectic points forms the boundary between the compartments. For example, the boundary between component B compartment and the component C compartment is the

10   trajectory of the eutectic points from E_2 to E_{SBC}. A straight line is used for simplification to illustrate the concept, however it should be understood that the actual boundary may be curved and can be calculated by commercially available thermodynamic model packages and then preferably is validated by use of experimental data obtained by routine experimentation within the teaching of the present invention.

15   When a solvent is used which has a melting temperature that is much lower than the melting temperature of both components A and B, the size of the solvent compartment in the projected phase diagram becomes very small as shown in Fig. 3. As the size of the solvent compartment becomes small, the binary eutectic points E_{SB} and E_{SA}, and the ternary eutectic points E_{SAC} and E_{SBC} move toward the apex of the triangle,

20   i.e. the pure solvent vertex. Hence in this case, a good approximation of the compartment size for components A, B and C is simply to draw the trajectory of the eutectic points from either E_1 or E_2 to the pure solvent point as shown in Fig. 4.

Of particular advantage, the inventors have discovered that a solvent may be employed to manipulate the location of the boundary of the various compartments, and thus the size of the compartments in the phase diagram as illustrated in Figs. 5 and 6. A solvent is provided in the solution, and the phase equilibrium behavior is selectively

5   controlled by controlling the composition and/or concentration of the solvent in the solution.

The solvent may be comprised of one component, or alternatively may be comprised of a binary mixture of two or more solvents components, or even a multi-component mixture and in each case is collectively referred to as a "solvent." Referring

10   specifically to Figure 5, a solvent (Solvent I) is employed. The Solvent I is selected such that the B compartment varies in size, in this case the size of compartment B becomes smaller, while the size of compartment A becomes larger. Thus, Solvent I produces a first phase behavior in the system. The compartments are complementary, and as one compartment becomes larger at least one of the other compartments becomes smaller due

15   to the interaction among components in the solution due to solubility and the like.

Fig. 6 shows how the size of the compartments will change upon selection of a different solvent. In this example, solvent II - which again may be a single solvent, a binary mixture or a multi-component mixture - is used. Here, the size of compartment B becomes larger while the size of compartment A becomes smaller. Thus, Solvent II

20   produces a second phase behavior in the system. This effect of the solvent on the size of the compartments is sometimes called a "swing" and can be clearly seen when the two phase diagrams are seen side by side.

As shown in the figures, when Solvent I is employed it produces the certain phase diagram due to the interaction among the components and Solvent I in the solution and their phase behavior which is governed by a variety of physical properties of the components such as solubility and the like. A different solvent, Solvent II produces a

5 different phase diagram again due to Solvent II's interaction among the components in the system.

This behavior allows one to manipulate the separation process to produce the desired compound or product from components in solution where the components can form one or more adducts according to the present invention. The effect of the solvent on

10 separation of the desired product(s) is now discussed in detail.

In one embodiment, when the solvent is comprised of a mixture of solvent components, the at least first and second solvent components exhibit at least first and second phase behaviors in the system that are diverse. Alternatively, the effect of the first and second solvent components on the phase behavior of the system may be closer,

15 or similar, to each other.

When characterized in terms of binary solubility, in particular the binary solubility of the compound of interest in the solvent component, the first and second solvent components exhibit solubilities that are preferably opposite; that is, one of the solvent components exhibits a higher solubility for a given component relative to the

20 other solvent component which exhibits a lower solubility for the component. Alternatively, the solubilities of the given component in the first and second solvent components may be closer to each other. However, to provide the greatest flexibility in

the operation of the process, it is preferable to employ solvent components which provide the more diverse phase behavior in the system.

Referring to FIGs. 5 and 6, manipulation of the phase equilibrium using two or more solvents with different phase behavior is shown according to two embodiments of

5   the present invention. In general, nature, size and shape of the adduct C and A and B phase compartments are dependent upon the type of solvent present in the product solution, as well as the phase equilibrium behavior of the binary system consisting of components A and B without the solvent. For example, for the case where adduct C is an incongruently melting adduct, the adduct composition lies to the right of the phase

10   boundary between the adduct and pure component B, as shown in FIG. 7. The terms size or shape of the compartment is meant to mean the size of the compartment in the projection of the poly-thermal phase diagram as shown in FIGs. 5 – 7, and not the complete poly-thermal phase diagram. As illustrated, the two phase compartments are complementary, that is when the A compartment increases the adduct C and/or B

15   compartment(s) decreases.

According to one preferred embodiment of the invention, Solvent I and Solvent II are selected such that the boundary by Solvent I from E_1 to S and the boundary by Solvent II from E_2 to S are overlapping. In other words, the compartment of component A when using solvent I, overlaps with the compartment of component B when using

20   solvent II. This overlapping is [VK1]shown on the phase diagram as indicated by the shaded area on FIG. 8. This overlap has a significant impact on the ability to selectively

control the separation of pure component A and/or pure component B from the adduct of components A and B.

Referring to FIG. 9, when Solvent I is used, the feed F is located in compartment A and the relevant boundary of compartment A is the line E_1PS (shown as a solid line).

5   Hence, solid A is recovered and the liquid composition is given by point P. In contrast, if Solvent II is used, the feed F is located in compartment B and the relevant boundary for compartment B is the line E_2QS (shown as a dashed line). Thus, solid B is recovered and the liquid composition is given by point Q as shown in the figure.

As stated above the phase behavior of the system results from the mutual

10   interaction of all the components present in the system; however, when selecting the solvent, the binary solubility data for the system components in various solvents may be used as a starting point to provide some indication of the type of phase behavior that may result for the complete system. The binary solubility data alone is not sufficient to determine the phase behavior of the complete system, however the binary solubility data

15   is a useful guideline for initial selection of the solvent component. Thus, the solvent is characterized most preferably in terms of its impact on the phase behavior of the system. Additionally or alternatively, the solvent may be characterized less reliably in terms of the relative binary solubility of the components in the solvent. When using solubility as the guideline for solvent selection, routine experimentation is performed within the

20   teaching of the present invention to evaluate the suitability of the selected solvent.

Given the teaching of the present invention, ternary phase equilibrium diagrams for different solvent compositions and/or concentration ratios may be created by those of

ordinary skill in the art without undue experimentation. For example, computer simulation programs and thermodynamic databases may be used to develop the approximate location of the boundary of the components and adduct compartments for different types of solvents and/or concentration ratios. As stated above the boundary

5   between the adduct compartment and the other compartments is the trajectory of eutectic points. The eutectic point is the location where both adduct and pure component solid coexist. The maximum recovery of the pure component is the maximum amount of component solid that can be recovered before adduct solid appears (or coexists with the component solid).

10   More specifically, the establishment of a thermodynamic model for the multi-component solid liquid equilibrium requires both theoretical calculation and experimental data for the model verification. Those of ordinary skill in the art can develop such thermodynamic models using known principles and tools.

The physical properties of the each component in the solution feed such as the

15   heat of fusion, melting point, and heat capacity are used to establish the thermodynamic model. The equilibrium relationship between the components and adduct as a function of temperature is needed. The binary interaction parameter among the components in the solution is either estimated or determined from experimental data. The theory for the activity coefficient is well developed, and a computer software program providing

20   numerical methods for solving simultaneous non-linear equations is typically used. Examples of suitable commercially available software programs for thermodynamic model establishment include PropertiesPlus from Aspen Technology Inc., HYPROP III

from Hyprotech, and ProPred from CAPEC at the Technical University of Denmark. The thermodynamic model is for the calculation of the concentration of the component when it is saturated and is a function of temperature and concentration. Since the effect of pressure on the saturation is not very significant in the pressure range considered in

5   typical separation units, the pressure effect is not taken into account in this thermodynamic model; however, the analysis may be extended to consider the pressure effect if desired.

Of significant advantage, a process can be designed to separate an adduct so as to recover one or both the pure components A and B by use of a "swing" of the solvent

10   effect on phase behavior between Solvent I and Solvent II according to the present invention.

The method of the present invention is carried out in a separation system which employs crystallization. According to one embodiment of the present invention, one exemplary embodiment of a separation system 5 which recovers both the pure

15   components A and B, is illustrated in FIG. 10. The separation system 5 generally includes two crystallizer units 10 and 12, liquid/solid separators 14 and 16, and at least one distillation column 18. The separation system 5 is generally downstream of a reactor system (not shown) and the solution conveyed to the separation system 5 is a product stream from the reactor system. The corresponding steam compositions are shown on the

20   phase diagram is shown in FIG. 11.

In the exemplary embodiment, feed F at Stream 1 (an adduct of A and B) is mixed with a recycle stream (Stream 6) to form a solution stream (Stream 2). This mixing is

illustrated in the projected poly-thermal phase diagram, as shown by the straight in FIG. 11. In this example Stream 6 contains components A, B and a solvent whose composition is termed as Solvent I.

Since the feed F is located in compartment A, solid product A is crystallized in crystallization unit 10 and then recovered from a solid/liquid separation device 14 as shown in the process block diagram of FIG. 10. The liquid from the solid/liquid separation device is Stream 3 and its composition is expressed on the projection of the poly-thermal phase diagram.

Stream 3 is mixed with the overhead of the distillation column 18 to form Stream 4. Stream 4 contains components A, B, and a solvent whose composition is termed Solvent II. Stream 4 is located in compartment B as shown in FIG. 11. Thus, pure solid B is crystallized in the crystallizer unit 12 and then is recovered from the solid/liquid separation device 16, as shown in the process block diagram of FIG. 10. It should be noted that Stream 3 is under the Solvent I environment. When mixed with the overhead of the distillation column 18, the solvent composition changes so as to give Stream 4 under Solvent environment II. Correspondingly, the phase boundary shifts from that indicated by the solid line to the one indicated by the dashed line in FIG. 11.

Conversely, when the overhead from the distillation column is separated from Stream 5 which is under the Solvent II environment, the resulting stream (Stream 6) is under the Solvent I environment. Correspondingly, the applicable phase boundary changes from the dashed line to the solid line in going from Stream 5 to Stream 6.

As described above the present invention provides for selectively controlling the phase equilibrium regions or compartments during crystallization by adjusting the composition of the solvent present in the solution to form a feed solution to a crystallizer having a certain feed composition such that either adduct or substantially pure

5   compounds of interest are produced. It is important to note that although the feed solution to a crystallizer is comprised of a multitude of components, including unreacted reactants and byproducts of the reaction, for process synthesis purposes it is sufficient to consider only the principal components. As such, the present invention provides a powerful tool allowing one to selectively operate the system to generate the desired phase

10   equilibrium region and thus crystallize the desired compound.

According to another embodiment of the present invention, an example of another separation system 9, which recovers the adduct in an intermediate step, and the single pure component desired from the separation process in another crystallization step, is shown in FIG. 12. Separation system 9 usually includes two crystallizer units 8 and 10, a

15   dissolver unit 13, liquid/solid separator units 15 and 17, and at least one distillation column 19. The separation system is generally downstream of a reactor system (not shown) and the solution conveyed to the separation system is a product stream from the reactor system. A part of the mother liquor from the separation system may be recycled to the reactor in order to increase the yield of the desired product. The stream compositions

20   for the separation process corresponding to FIG. 12 are shown on the projected polythermal phase diagram in FIG. 13.

In this exemplary embodiment, components A and B exhibit an incongruently melting adduct, and component B is the final desired component from the separation. A phase diagram of this system in presence of a Solvent was shown earlier in FIG. 7. The incongruent melting point (E_2) is the point where the adduct melts, and the liquid

5   composition and the solid composition are not the same. In the absence of solvent, when the adduct C melts, the liquid composition is at point E_2 while some solid B is formed as well. This is why the location of the adduct C is between point E_2 and pure B. In presence of a solvent, the boundary between the pure B and the pure adduct C compartment is given be the trajectory of the incongruent melting points at different

10   solvent contents. This is shown by the line from the E_2 to the Solvent vertex in FIG. 7.

Referring specifically to FIG. 12, in this exemplary embodiment, feed F at Stream 1 is mixed with a recycle stream (Stream 6) to form a solution stream (Stream 2). This mixing is illustrated in the projected poly-thermal phase diagram, as shown by the straight in FIG. 13. In this example Stream 6 contains components A, B and Solvent I.

15   Since Stream 2 is located in the adduct C compartment, solid adduct is crystallized in crystallization unit 8 and then recovered from a solid/liquid separation device 15 as shown in the process block diagram of FIG. 12. The liquid from the solid/liquid separation device is Stream 4 and its composition is expressed on the projection of the poly-thermal phase diagram.

20   Stream 3 which is solid adduct C is dissolved in dissolver 13 using the overhead of the distillation column 19 to form Stream 5. Stream 5, containing components A, B, and Solvent I, is located in compartment B as shown in FIG. 13. Thus, pure solid B is

crystallized in the crystallizer unit 10 and then is recovered from the solid/liquid separation device 17, as shown in the process block diagram of FIG. 12. The bottoms from the distillation column 19 is rich in the undesired component A and is recycled to the reactor where part of it gets converted to the desired component B.

5   While two examples of a separation system are shown herein for illustration purposes, the method of the present invention may be carried out in other types of crystallization separation systems.

To selectively control the location of the composition in the solution at the feed to the crystallizer, the composition of the various components in the solution may be

10   adjusted to certain amounts (typically weight or mol percent) to place the feed composition in a crystallizer at a location selectively within a desired region of the phase diagram. The composition of the solution at the feed to a crystallizer may be adjusted in a variety of ways. Any one of, or combination of, the components A and B in solution, the solvent, or mixture of solvent components may be added to, or removed, from the

15   solution prior to the crystallizer to adjust the feed composition to the desired phase compartment. Alternatively, the composition of reactants fed to a reactor and the reaction conditions in terms of temperature, conversion, and the like may be adjusted to provide the desired composition in the solution from the reactor. For example, the flow rate of reactants to the reactor may be varied to provide a certain weight or mol percent

20   of component A or B in the product solution.

While certain embodiments have been described herein, it should be understood to those of ordinary skill in the art that the method of the present invention is

applicable to any solid and liquid system with an adduct, exhibiting either a congruent melting point or an incongruent melting point. Further, there may be present one, two or more eutectic points, and there can be multiple adducts formed.

5   Experimental

Simulated experiments are provided below to further illustrate the system and method of the present invention. These simulated experiments are provided for illustration purposes only and are not intended to limit the scope of the invention in any way.

10   **Example 1**

This example refers to one of the processes of recovering both pure components A and B from their adduct C, by use of a "swing" of the solvent effect on phase behavior between Solvent I and Solvent II according to the present invention. This embodiment of the invention was described earlier and illustrated in FIGs. 10 and 11.

15   Solvent I and Solvent II are selected such that the boundary by Solvent I from E_1 to S and the boundary by Solvent II from E_2 to S are overlapping. In other words, the compartment of component A when using solvent I, overlaps with the compartment of component B when using solvent II.

In this particular example, the Solvents I and II are both comprised of a mixture of

20   two solvent components, S_1 and S_2. S_1 and S_2 are selected such that the compartment of component A when using S_1, overlaps with the compartment of component B when using S_2.

For a given solvent combination (S_1 and S_2), the maximum overlap possible is fixed. The actual overlap produced in the process (less than the maximum overlap) is decided by the actual compositions of solvent (mixture) I and solvent (mixture) II in the crystallizers 10 and 12, respectively.

5   Referring to FIG. 10, the amount and composition of solvent removed by the solvent recovery unit 18 will determine the extent of "swing" between Solvent I environment and Solvent II environment. For a lower solvent flow or a smaller composition change, the swing, and hence the overlap will be less, while for a larger solvent flow or a larger composition change, the overlap will be more.

10   This example shows how the swing in the phase boundary changes as the solvent composition in the crystallizers 10 and 12 changes. The phase boundaries resulting from using different Solvent compositions in the crystallizer units 10 and 12 are shown on the phase diagram in the FIG. 14. The dark lines refer to the boundary between the component B compartment and the Adduct C compartment, whereas the light lines refer

15   to the boundary between the component A compartment and the Adduct compartment. Three cases are considered.

Case 1:

          When the Solvent I and II environments refer to pure S_1 and pure S_2, the resulting

20   phase boundaries are dotted lines as shown in FIG. 14, with a corresponding large overlap.

Case 2:

The solvent composition in the crystallizer unit 10 (Solvent I environment) is 80% S_1 and 20% S_2, while the solvent composition in the crystallizer unit 12 (Solvent II environment) is 20% S_1 and 80% S_2. The phase boundaries are given by dashed lines,

5 with a lesser overlap.

Case 3:

The solvent composition in the crystallizer unit 10 (Solvent I environment) is 60% S_1 and 40% S_2, while the solvent composition in the crystallizer unit 12 (Solvent II

10 environment) is 40% S_1 and 60% S_2. The phase boundaries are given by solid lines, resulting in no overlap between Compartment A and Compartment B. This will make the process infeasible.

Example 2

15 This example examines in more detail, the system considered in Example 1. Although the overlap in the phase boundaries leads to a feasible process as discussed above, the extent of overlap has a large effect on the efficiency of the process. A smaller overlap leads to lower per pass recoveries of pure A and B in the crystallizers. This leads to a larger flow of the recycle mother liquor stream (Stream 6).

20 This example considers the effect of the solvent flowrate on the extent of overlap, the recoveries in the crystallizer, and the recycle flow, for an imaginary chemical system. The feed to the process is considered to be the equimolar adduct C of A and B, with a

flow rate of 100 kmol/h. This is because starting from the adduct usually leads to the most difficulty in separation. Similar process configurations can be devised to separate a feed with a different composition of A and B than the adduct composition.

Case 1 considers a small swing in the solvent compositions of the two crystallizer

5   units, 10 and 12. This is achieved by having a low solvent recycle flow (such as 60 kmol/h) from the top of the distillation column 18. The small swing in the solvent environments results in a small overlap between the component A and component B compartments, as shown in FIG. 15. Due to the small overlap, the region of feasible process operation is smaller, and hence a lower per pass recovery is achieved in the

10   crystallizers 10 and 12. Since the separation duty on the process remains the same, this leads to a large recycle flow (Stream 6). Case 2 and 3 show how a larger overlap can lead to a lower recycle and hence a more efficient process. FIGs. 16 and 17 show the corresponding stream compositions and the phase diagrams for Case 2 and 3. Table 1 below, summarizes the per pass recoveries in the crystallizers and the recycle flows for

15   all three cases of the simulated experiments.

TABLE 1

Case	Per pass recovery (%)		Solvent Recycle	Mother Liquor Recycle (Stream 6)
	A	B	(kmol/h)	(kmol/h)
1: "Small Overlap"	24.4	53.6	60	360
2: "Medium Overlap"	39.4	72.2	100	160

3: "Large Overlap"	46.2	79.1	140	110

Example 3

This example examines a system where the components A and B exhibit an

5   incongruently melting adduct, and component B is the final desired component from the

reaction – separation. This embodiment of the invention was described earlier and

illustrated in FIGs. 12 and 13. One exemplary embodiment of the process for separating

the desired component B from the mixture of A, B and solvent is illustrated in FIG. 12.

The reactor converts a reactor feed rich in undesired component A but with a low

10   concentration of desired component B, into a stream with a higher concentration of

component B. This is the feed to the separation system (Stream 1). The separation

consists of first recovering the incongruently melting adduct C in crystallizer 8. This is

then dissolved in the dissolver unit 13 using a suitable solvent so as to place the

composition of the resulting stream (Stream 5) in the component B compartment. Pure

15   component B is then recovered in crystallizer unit 10.

The choice of Solvent used has a great impact on the location of the phase

boundaries shown in FIG. 13. This in turn greatly affects the efficiency for the separation

of the adduct C and the pure component B in the process described above.

Although a single solvent is sufficient to give a feasible separation process, this

20   example illustrates how the efficiency of separation can be enhanced by using a "swing"

in the solvent composition between Solvent I and Solvent II in the two crystallizer units 8

and 10, so as to achieve phase boundaries which are advantageous for the particular

separation being carried out in each crystallizer.

For the above process, the process streams are identified on the projection of the

polythermal phase diagram in FIGs. 18 and 19, for two different solvent environments, I

5   and II. For solvent I, the phase boundaries curve in a fashion so as to give a large

compartment for B, while the compartment for the adduct C is small. On the other hand,

for Solvent II, the phase boundaries are such that while the crystallization compartment

for the Adduct C is large, that for component B is small. This leads to higher per pass

recovery for component B for the case of Solvent I, as compared to Solvent II, and higher

10   per pass recoveries for the adduct C for the case of Solvent II as compared to Solvent I.

In this situation, we may operate the process advantageously using a solvent

swing, such that the crystallizer for component B is under the Solvent I environment,

while the adduct C crystallizer is under the solvent II compartment. The process streams

for such an operation are identified on the projection of the polythermal phase diagram in

15   FIG. 20. By thus "swinging" the solvent composition from one solvent to the other, better

per-pass recovery for both component B and the adduct C are obtained than were possible

using the individual solvents alone. Simulated examples of this effect are shown in Table

2 below.

20   TABLE 2

Case	Per pass recovery of Adduct, C (%)	Per pass recovery of B (%)
1: Solvent I	52.7	85.0
2: Solvent II	65.8	22.9
3: Solvent I/II	63.5	84.2

The foregoing description of specific embodiments and examples of the invention have been presented for the purpose of illustration and description, and although the invention has been illustrated by certain of the preceding examples, it is not to be construed as being limited thereby. They are not intended to be exhaustive or to limit the invention to the precise forms disclosed, and obviously many modifications, embodiments, and variations are possible in light of the above teaching. It is intended that the scope of the invention encompass the generic area as herein disclosed, and by the claims appended hereto and their equivalents.